

# The effect on photochemical smog of converting the U.S. fleet of gasoline vehicles to modern diesel vehicles

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[1] With the increased use of particle traps and nitrogen oxide ( $\text{NO}_x$ ) control devices to reduce air pollution, “modern” diesel vehicles are being encouraged over gasoline vehicles globally as a central method of slowing global warming. Data to date, though, suggest that the  $\text{NO}_2$ : $\text{NO}$  ratio from modern diesel may exceed that of gasoline, and it is difficult to reduce diesel  $\text{NO}_x$  below gasoline  $\text{NO}_x$  without increasing particle emissions. Here, it is calculated that, unless the diesel  $\text{NO}_2$ : $\text{NO}$  ratio and total  $\text{NO}_x$  are reduced to those of gasoline, modern diesel, which should have lower hydrocarbon (HC) and carbon monoxide (CO) emissions than gasoline, may still enhance photochemical smog at the surface and aloft, on average, over the U.S. relative to gasoline. The reason is that vehicle-produced smog in the U.S. depends more on  $\text{NO}_x$  and the  $\text{NO}_2$ : $\text{NO}$  ratio than on HCs or CO. It is also found that vehicle  $\text{NO}_x$  controls may be more effective than  $\text{NO}_2$ : $\text{NO}$  ratio controls at reducing ozone.

**INDEX TERMS:** 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0317 Atmospheric Composition and Structure: Chemical kinetic and photochemical properties; 0322 Atmospheric Composition and Structure: Constituent sources and sinks; 0345 Atmospheric Composition and Structure: Pollution—urban and regional (0305); 0365 Atmospheric Composition and Structure: Troposphere—composition and chemistry. **Citation:** Jacobson, M. Z., J. H. Seinfeld, G. R. Carmichael, and D. G. Streets (2004), The effect on photochemical smog of converting the U.S. fleet of gasoline vehicles to modern diesel vehicles, *Geophys. Res. Lett.*, 31, L02116, doi:10.1029/2003GL018448.

## 1. Introduction

[2] Many countries are developing strategies to slow global warming. One strategy already being implemented in much of the European Union and considered in California is to encourage diesel over gasoline vehicles because diesel vehicles are thought to emit less  $\text{CO}_2$  (see auxiliary material<sup>1</sup>) and “modern” diesel vehicles (those with particle traps and, potentially,  $\text{NO}_x$  control devices) are cleaner than diesel vehicles of the past. Despite this widespread belief, no study has examined the potential effects on air pollution of replacing gasoline with modern diesel vehicles. Given

that changes in air pollution can affect health, such a study is warranted.

[3] Table 1 compares estimated emissions of several pollutants from modern gasoline and diesel passenger vehicles, where the diesel contains a particle trap but no  $\text{NO}_x$  device. In the absence of a trap (not shown), diesel CO and hydrocarbon (HC) emissions are similar to those from gasoline. A trap added to a diesel not only collects then burns off particles (during regeneration); it also catalyzes the oxidation of CO to  $\text{CO}_2$  and HCs to  $\text{CO}_2$  and  $\text{H}_2\text{O}$ .

[4] Diesel vehicles, with or without a trap and without a  $\text{NO}_x$  device, emit 4–30 times more  $\text{NO}_x$  than do gasoline vehicles (Table 1). Under California Low Emission Vehicle II emission standards (LEV II),  $\text{NO}_x$  emissions from passenger cars and light-duty vehicles must be  $<50$  mg/mi (31 mg/km) and particle emissions must be  $<10$  mg/mi (6.2 mg/km) by 2004. These are the toughest proposed standards worldwide. Currently, many new gasoline vehicles meet both standards and others are close (e.g., Table 1). Table 1 suggests that, with the addition of a particle trap, diesel may meet the particle standard but not the  $\text{NO}_x$  standard. As such, technologies to reduce diesel  $\text{NO}_x$  (and gasoline  $\text{NO}_x$  further) are being improved. A tradeoff, though, exists between  $\text{NO}_x$  and particle reductions in diesel [Johnson, 2001] such that although diesel  $\text{NO}_x$  emissions will decrease, they may or may not drop to those of gasoline because of the need to ensure maximum particle controls. Here, it is assumed that all diesel vehicles will have a  $\text{NO}_x$  device, reducing diesel  $\text{NO}_x$  emission to or nearly to that of gasoline.

[5] Another issue with modern diesel is the  $\text{NO}_2$ : $\text{NO}$  ratio. In diesel vehicles without a trap and gasoline vehicles, the ratio is about 10:90 [Ayala *et al.*, 2002; Ullman *et al.*, 2002]. In Europe, many gasoline passenger vehicles emit about 4%  $\text{NO}_2$  whereas light-duty trucks may emit about 18%  $\text{NO}_2$  [Soltic and Weilenmann, 2002] again suggesting that a reasonable fleet-average of 10:90. In heavy-duty diesel vehicles with a particle trap, NO must be converted to  $\text{NO}_2$ , which oxidizes soot in the trap. The resulting  $\text{NO}_x$  exhaust contains 30–70%  $\text{NO}_2$  [Ayala *et al.*, 2002; Ullman *et al.*, 2002; Guo *et al.*, 2003]. Some passenger vehicles with a trap use a catalyst, such as cerium, instead of  $\text{NO}_2$ , for oxidizing soot. In the exhaust of such vehicles, though,  $\text{NO}_2$  is still 13–39% of  $\text{NO}_x$  [PSA Peugeot-Citroen, pers. comm., 2002]. In sum, diesel vehicles with a trap and without an  $\text{NO}_x$  device emit more  $\text{NO}_x$  and a higher ratio of  $\text{NO}_2$ : $\text{NO}$  than do gasoline vehicles.

[6] Here, the possible effects of replacing gasoline with diesel vehicles are examined when  $\text{NO}_x$  emissions are 0, 25,

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**Table 1.** Emissions from a diesel vehicle with a particle trap and without an NO<sub>x</sub> device compared with those from a gasoline vehicle

	CO (g/km)	HCs (g/km)	NO <sub>x</sub> (g/km)	% NO <sub>x</sub> as NO <sub>2</sub>	PM (mg/km)
Diesel, w/trap	0.10 <sup>a</sup>	0.030 <sup>a</sup>	0.34 <sup>a</sup> , 0.3–0.4 <sup>b</sup>	13–39 <sup>b</sup> 30–70 <sup>c,d,e</sup>	1.5–15 <sup>a,g,h,i,j</sup>
Gasoline	0.26 <sup>a</sup>	0.059 <sup>a</sup>	0.042 <sup>a</sup> , 0.01–0.1 <sup>h</sup>	10 <sup>c,d,e</sup> 4.3–17.6 <sup>f</sup>	0.05–2 <sup>i,k</sup>

This study assumes an NO<sub>x</sub> device will be added to diesel, reducing NO<sub>x</sub> emissions to 0–100% higher than those of gasoline.

<sup>a</sup>Ahlvik [2002] citing Peugeot 307 HDi FAP diesel and Peugeot 307 1.6 110 hp gasoline certification data.

<sup>b</sup>PSA Peugeot-Citroen, *pers. comm.* [2002].

<sup>c</sup>Ayala *et al.* [2002].

<sup>d</sup>Guo *et al.* [2003].

<sup>e</sup>Ullman *et al.* [2002].

<sup>f</sup>Soltic and Weilenmann [2002] (4.3% for passenger vehicles; 17.6% for light-duty trucks).

<sup>g</sup>Salvat *et al.* [2000].

<sup>h</sup>Durbin and Norbeck [2002].

<sup>i</sup>ACEA [2002].

<sup>j</sup>Jeuland *et al.* [2002].

<sup>k</sup>Maricq *et al.* [1999].

50, and 100% higher for diesel than for gasoline and NO<sub>2</sub>:NO ratios are 10:90, 20:80, and 30:70 for diesel and 10:90 for gasoline. In all cases, modern diesel is assumed to emit 50% less HCs and CO and twice as much primary particle mass (Table 1). Since HC and CO emissions from diesel are assumed to be less than those of gasoline, it is assumed that diesel will pollute less than gasoline if its NO<sub>x</sub> emission falls below that of gasoline.

[7] This study was carried out with GATOR-GCMOM, a nested global-to-urban pollution/weather/climate model [Jacobson, 1997a, 1997b, 2001a, 2001b, 2001c, 2001d, 2002]. It treated time-dependent gas, aerosol, radiative, dynamical, cloud, land, and ocean processes over two layers of nesting, the globe and the U.S. The U.S. resolution was 0.5°S–N × 0.75°W–E (about 55 km S–N × 68 km W–E), so cities were not resolved. The relatively coarse inner grid allows the whole U.S. to be treated but increases uncertainty, some of which is addressed in a previous study with nested grids [Jacobson, 2001b, Table 2].

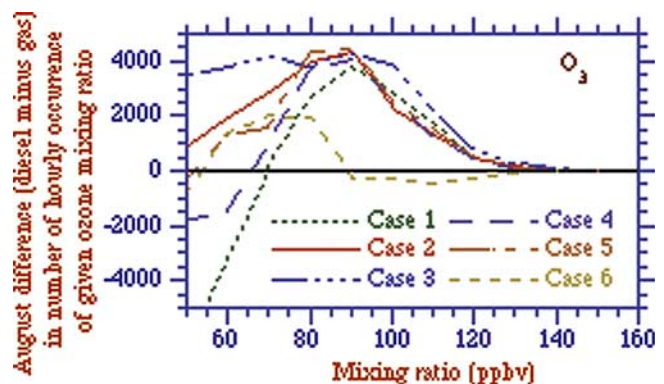
[8] Gridded, hourly anthropogenic emissions of speciated total organic gas (TOG), NO<sub>x</sub>, SO<sub>x</sub>, CO, NH<sub>3</sub>, PM<sub>2.5</sub>, and PM<sub>10</sub> for the U.S. and parts of Canada/Mexico were obtained from the 1999 U.S. National Emission Inventory [USEPA, 2003]. The inventory accounts for over 370,000 stack and fugitive sources, 250,000 area sources, and 1700 source classification code (SCC) categories of on- and nonroad mobile sources (including 837 categories of gasoline vehicles: motorcycles, cars, trucks, recreational/gasoline vehicles: motorcycles, cars, trucks, construction/farm/industrial vehicles, etc.). Yearly U.S. emissions of NO<sub>x</sub>, CO, and TOG from gasoline-only mobile onroad plus nonroad sources in the inventory were  $4.06 \times 10^6$ ,  $7.92 \times 10^7$ , and  $1.09 \times 10^7$  metric tonnes/yr, respectively, representing 19.2%, 73.0%, and 37.0% of all U.S. anthropogenic emissions of these pollutants in the inventory. Other mobile-sources (mostly diesel) accounted for another 35.0% of NO<sub>x</sub> but lower levels of other pollutants. The model also included biogenic emission of isoprene, monoterpenes, VOCs, and NO. The auxiliary material describes processes in the model and its initialization and compares model results with measurements.

## 2. Simulations and Results

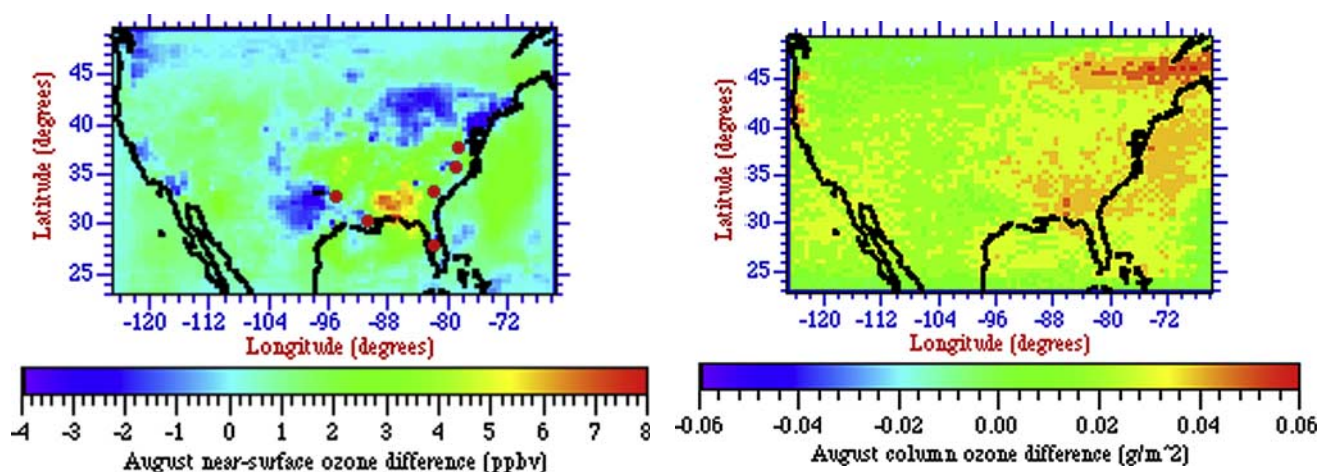
[9] A baseline and six sensitivity simulations, in which all SCC categories of gasoline vehicles were switched to

diesel (as described in Figure 1), were run for August and February, 1999. Figure 1 shows the August difference in the frequency distribution of hourly near-surface O<sub>3</sub> over the U.S. for the six cases. The switch to diesel increased O<sub>3</sub> > 60 ppbv in all six cases. The number of increases in case 2 (50% NO<sub>x</sub> increase) was between those of cases 1 (25% NO<sub>x</sub> increase) and 3 (100% NO<sub>x</sub> increase). Changes in NO<sub>2</sub>:NO at the same NO<sub>x</sub> level (cases 2, 4, 5) had less effect on O<sub>3</sub> than did changes in total NO<sub>x</sub>. For case 3, diesel increased O<sub>3</sub> > 120 ppbv by 51% and >80 ppbv by 17%.

[10] In case 6, ozone increased when NO<sub>2</sub>:NO increased from 10:90 to 20:80 with no change in NO<sub>x</sub>. This occurred



**Figure 1.** Difference (diesel minus gasoline) in the number of occurrences of different ozone mixing ratios (in 10 ppbv increments), summed over every hour of August and over every near-surface regional-domain (U.S.) grid cell, for six cases. Case 1: diesel:gasoline NO<sub>x</sub> = 1.25 and diesel NO<sub>2</sub>:NO = 20:80; Case 2: diesel:gasoline NO<sub>x</sub> = 1.5 and diesel NO<sub>2</sub>:NO = 20:80; Case 3: diesel:gasoline NO<sub>x</sub> = 2.0 and diesel NO<sub>2</sub>:NO = 20:80; Case 4: diesel:gasoline NO<sub>x</sub> = 1.5 and diesel NO<sub>2</sub>:NO = 10:90; Case 5: diesel:gasoline NO<sub>x</sub> = 1.5 and diesel NO<sub>2</sub>:NO = 30:70; Case 6: diesel:gasoline NO<sub>x</sub> = 1.0 and diesel NO<sub>2</sub>:NO = 20:80. In all cases, gasoline NO<sub>2</sub>:NO = 10:90, diesel CO and primary HC emissions equaled half those of gasoline (Table 1), and diesel primary particle emissions were twice those of gasoline (a low estimate from Table 1). The summed number of occurrences of all ozone mixing ratios (down to 0–10 ppbv) was equal in all cases.



**Figure 2.** Modeled August difference (averaged over every hour in August) in (a) near surface  $O_3$  and (b) column  $O_3$  from case 2 of Figure 1. Red dots are locations for comparisons with data in the Supplementary Information.

because, when  $NO_x$  is emitted continuously as  $NO_2$ ,  $O_3$  forms almost directly by  $NO_2 + UV \text{ light} \rightarrow NO + O$  followed by  $O + O_2 + M \rightarrow O_3 + M$ . When the same  $NO_x$  is emitted as  $NO$ , the  $NO$  must first react with  $O_3$  or peroxy radicals to form  $NO_2$  before creating new  $O_3$ , so this path destroys some of the  $O_3$  that is created. There was little change in  $O_3$  at high mixing ratios in case 6 possibly because additional  $O_3$  formed by emitting  $NO_x$  as  $NO_2$  at high  $O_3$  was small relative to that formed by other means and/or because  $NO$  levels at high  $O_3$  were high enough to titrate the additional  $O_3$ .

[11] Statistics for PAN,  $NO_2$ ,  $HNO_3$ , particulate nitrate, and particulate BC also showed increases at all upper mixing ratios following the switch to diesel. Nitrate increases were 5–15 times greater than BC increases (which were small due to the small difference between diesel with a trap versus gasoline particulate emissions) and occurred primarily where  $HNO_3$  increased (see auxiliary material). Primary particulate organic matter (OM) increased slightly, but secondary OM decreased, as expected. Statistics for CO and most primary HCs showed decreases at upper mixing ratios. Although overall mixing ratios of  $O_3$  were lower in February than August, trends for February were similar to those for August. For example, for case 2 in February, diesel increased  $O_3$  at all mixing ratios  $>45$  ppbv.

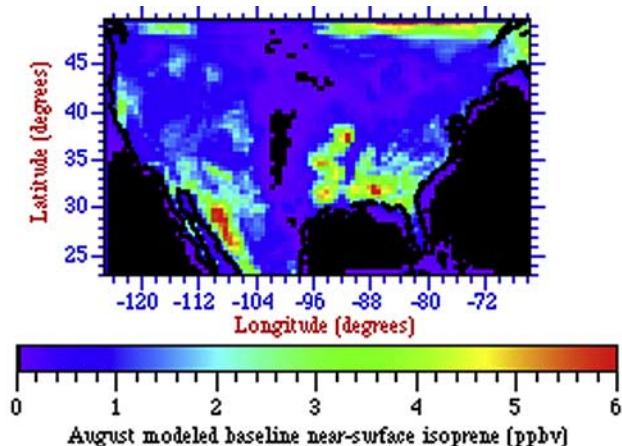
[12] Figure 2a shows the August-average U.S. difference in  $O_3$  following conversion to diesel in case 2. Ozone increased in more than 75% of the U.S. Most increases occurred in areas of high average  $O_3$ . The maximum increase (8 ppbv, in the Southeast) was twice the maximum decrease (4 ppbv, along the northeast corridor) and four times the decrease in Los Angeles (2 ppbv). Other  $O_3$  decreases occurred in parts of the north-Midwest and Great Plains, where  $O_3$  was relatively low. For these reasons, diesel increased the number of hours of high  $O_3$  over the U.S. (Figure 1). At about 20% of Northeast locations where  $O_3$  decreased overall, it increased in the afternoon, during its peak, but decreased more at night, when  $O_3$  was low. In remaining decrease cases,  $O_3$  decreased during day and night.

[13] Changes in  $O_3$  in Figure 2a can be explained partly in terms of an ozone isopleth [e.g., Seinfeld and Pandis, 1998]. Increases in  $O_3$  were caused by increases in  $NO_x$  in the

presence of high total HCs, as expected from an isopleth. Although diesel reduced vehicle HCs, total HCs were high in the Southeast because biogenic HC emissions were high there (Figure 3). Decreases in  $O_3$  were due to three factors: (1) in urban areas (e.g., Los Angeles, northeast corridor), increases in  $NO_x$  at high  $NO_x$  and at low biogenic HCs decreased  $O_3$ , (2) in areas of low biogenic HCs and moderate to high  $NO_x$ , reductions in vehicle HCs due to diesel decreased  $O_3$  production, and (3) in areas of high nighttime  $NO_x$ , titration by excess  $NO_x$  decreased nighttime  $O_3$ . In nearly every area where  $O_3$  decreased from diesel, biogenic HC emissions were low (compare Figures 2a and 3).

[14] Although near-surface  $O_3$  decreased slightly in some places due to diesel,  $NO_2$  increased in those locations (Supp. Info), and the number of increases in high- $O_3$  mixing ratios far outweighed the number of decreases (Figure 1). Ozone also increased in several grid cells containing non-attainment areas (e.g., Birmingham, Atlanta, Baton Rouge, St. Louis, Dallas-Fort Worth, Houston-Galveston-Brazoria) and areas close to nonattainment [Chameides *et al.*, 1997] (the model did not resolve the urban areas themselves).

[15] Figure 2b shows that diesel increased ozone column abundance across the entire U.S. Excess column  $O_3$  can be



**Figure 3.** August baseline monthly-averaged modeled near-surface isoprene.



transported long distances and contributes to global warming. Excess  $O_3$  aloft from diesel was either generated at the surface and convected aloft or generated from excess  $NO_x$  that escaped the boundary layer and reacted with background HCs and their products aloft. Winds aloft moved excess  $O_3$  aloft faster than at the surface, increasing column  $O_3$  over a wide area, including over areas where surface  $O_3$  decreased.

[16] Diesel increased near-surface  $NO_2$ , PAN, and  $HNO_3$  on average (auxiliary material)<sup>1</sup>. Since  $NO_2$  and  $HNO_3$  can only increase whereas  $O_3$  can increase or decrease with an increase in  $NO_x$ ,  $NO_2$  and  $HNO_3$  decreased in fewer areas than did  $O_3$ . Increases in  $HNO_3$  are important because  $HNO_3$  forms particles, which are known health hazards. Locations of PAN changes were similar to those of  $O_3$  changes since PAN, like  $O_3$ , depends on HCs and  $NO_x$ . Mixing ratios of primary organics (e.g., primary aromatics, alkanes) decreased over the U.S. following the switch to diesel, as expected.

[17] The results here agree with those from several studies that have found that areas of high natural HC emission may be  $NO_x$  limited whereas urban areas (e.g., the northeast corridor, Los Angeles) may be HC limited [e.g., *Trainer et al.*, 1987; *Sillman et al.*, 1990; *NRC*, 1991; *Roselle et al.*, 1991; *McKeen et al.*, 1991; *Jacob et al.*, 1993; *Fiore et al.*, 1998; *Streets et al.*, 2001]. Here, it is similarly found that vehicle  $NO_x$  emission controls may be more effective than vehicle HC controls at reducing  $O_3$  over most of the U.S., particularly in the Southeast, where biogenic emissions are high. An exception may be Los Angeles and along the Boston-Washington corridor, where natural HC emissions are low. This result is particularly consistent with that of *Jacob et al.* [1993] who modeled  $O_3$  over the U.S. with resolution eight times coarser but found that a 50% decrease in  $NO_x$  decreased summer  $O_3$  over the mid-southeastern U.S. by about 10–15 ppbv and slightly increased  $O_3$  along the northeast coast, whereas a decrease in HCs of 50% decreased  $O_3$  by only 3 ppbv.

[18] Previous studies have not examined the effect of  $NO_x$  versus HCs on column  $O_3$  or pollutants aside from  $O_3$  across the U.S. or the effect of gasoline vehicle  $NO_x$  (as opposed to total  $NO_x$ ) versus HC emission controls on near-surface  $O_3$ . The difference between gasoline vehicle emission and total emission is that the mass ratio of gasoline  $NO_x$ :HC in the U.S. is about 0.34:1 whereas that of total anthropogenic  $NO_x$ :HC is about 0.72:1. Finally, previous studies have not examined the effect of the  $NO_2$ :NO ratio on air pollution across the U.S. Here it was found that reducing the ratio may slightly reduce  $O_3$ , on average, but not so much as reducing  $NO_x$ .

### 3. Conclusion

[19] This study concludes that the replacement of gasoline with modern diesel vehicles in the U.S. may drive up photochemical smog, including total column ozone, near-surface ozone, and nitrogen-containing species, over the U.S. on average and in the Southeast in particular, unless  $NO_x$  emissions and the  $NO_2$ :NO ratio from diesel vehicles are reduced to or below those of gasoline vehicles. The study also finds that vehicle  $NO_x$  controls may be more effective than  $NO_2$ :NO ratio controls at reducing ozone. The

results apply even though modern diesels emit less HCs and CO than gasoline vehicles because, on average, photochemical smog depends more on vehicle  $NO_x$  than hydrocarbon emissions. Thus, if gasoline vehicles meet the toughest planned  $NO_x$  regulations worldwide, and if diesel vehicles with a particle trap and  $NO_x$  device also meet such standards but emit  $NO_x$  at a slightly higher level, diesel vehicles may exacerbate smog relative to gasoline vehicles. To remedy this potential problem, technology is necessary to reduce  $NO_x$  and the  $NO_2$ :NO ratio of diesel to or below those of gasoline without increasing particulate matter emissions.

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